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RESEARCH PAPER

Rheological and mechanical properties of wood fiber-PP/PE blend composites

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Abstract: For evaluation of the rheological and mechanical properties of highly filled wood plastic composites (WPCs), polypropylene/polyethylene (PP/PE) blends were grafted with maleic anhydride (MAH) to enhance the interfacial adhesion between wood fiber and matrix. WPCs were prepared from wood fiber up to 60 wt.% and modified PP/PE was blended by extrusion. The rheological properties were studied by using dynamic measurement. According to the strain sweep test, the linear viscoelastic region of composites in the melt was determined. The result showed that the storage modulus was independent of the strain at low strain region (<0.1%). The frequency sweep results indicated that all composites exhibited shear thinning behavior, and both the storage modulus and complex viscosity of MAH modified composites were decreased comparing to those unmodified. Flexural properties and impact strength of the prepared WPCs were measured according to the relevant standard specifications. The flexural and impact strength of the manufactured composites significantly increased and reached a maximum when MAH dosage was 1.0 wt.%, whereas the flexural modulus after an initial decreased, also increased with MAH dosage. The increase in mechanical properties indicated that the presence of anhydride groups enhanced the interfacial adhesion between wood fiber and PP/PE blends.

Keywords: wood fiber; PP/PE blends; composites; rheological properties; mechanical properties

Introduction

In recent years, more and more scientists pay attention to the use of wood fiber as reinforcement filler in plastics. Wood fibers have several advantages over conventional reinforcing materials, such as abundance, low cost, low density, high specific strength and stiffness, non-abrasive to processing equipment and renewability (Bledzki et al. 1998; Zadorecki et al. 1989). Wood-plastic composites can be processed in conventional processing equipments for plastics, such as extrusion and injection moulding. The addition of wood increased the stiffness of the plastic matrix. When proper interfacial adhesion between the wood and the plastic matrix is achieved, the specific strength of the composites is also higher than unfilled plastics (Bengtsson et al. 2007).

In previous studies, the wood fiber content was generally from 10 to 40 wt.%. However, the main purpose for the addition of wood fiber to plastics is to reduce the cost and improve the stiffness, and it would be interested to study the potential of the wood fiber filling content (exceeding 50 wt.%). The poor compatibility between polar hydrophilic wood fibers and non-polar hydrophobic plastics becomes the primary problem, which results in poor mechanical properties of the final product (Karmarkar et al. 2006).

In order to achieve reasonable mechanical properties at such high fiber loadings, it is important to have a better dispersion of the fibers in the matrix, a more effective wetting of fibers by matrix resin and good adhesion between the two phases (Bengtsson et al. 2007; Felix et al. 1991). The interfacial adhesion can be improved by using coupling agents or surface modification of the fibers or the matrix. It had been conformed that the maleated polypropylene (MAPP) can significantly improve the interfacial adhesion between wood fiber and matrix (Hon et al. 2006). It is also reported that the mechanical properties can be improved by using silane or isocyanate coupling agents (Karmarkar et al. 2006; Karnani et al. 1997; Maldas et al. 1993).

In this study, polypropylene (PP) and high density polyethylene (HDPE) blends were used as matrix. In order to achieve reasonable mechanical properties at high fiber loadings, uniform dispersion of the wood fibers in the matrix and the good interfacial adhesion should be taken into account. Thus, the grafting reaction of MAH onto PP/PE blends proceeded in a co-rotating twin screw extruder. Composites of wood fiber and modified PP/PE blends were prepared by melt extrusion. The objectives of this study were to evaluate the rheological and mechanical properties of wood fiber-PP/PE composites. The small amplitude oscillation test was selected for investigating the viscoelastic

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behavior of the composites melt. The mechanical properties of the composites were measured according to the relevant standard specifications.

Material and methods

Materials

Polypropylene (T30S) and high density polyethylene (2200J) were provided by Daqing Petrochemical Company, China. Wood fiber (30–50 mesh) with 8% of moisture content was supplied by Harbin Yongxu Co. Ltd. All the other chemicals including Maleic anhydride (MAH) and dicumyl peroxide (DCP) were reagent-grade products and used without further purification.

Methods

Grafting procedure

Melt grafting of MAH onto PP/PE blends was performed in a co-rotating twin screw extruder in the presence of dicumyl peroxide (DCP) as initiator. Before extruding, MAH and DCP were mulled to disperse homogeneously and then mixed with PP/PE (80/20) blend granules. According to previous studies, a fixed ratio of MAH to DCP (10:1) was employed. The reaction temperature was 190 °C, and the rotation speed of the screws was 50 rpm. A series of grafting products with 0.5, 1.0 and 1.5 wt.% of MAH (M0.5, M1.0 and M1.5) were prepared respectively. The extruded products were cooled at ambient temperature and granulated.

Preparation of composites

Wood fibers were dried at 105°C for 8 h to the moisture content below 1% before the extrusion process. Wood fiber (60 wt.%) and PP/PE blends (40 wt.%, un-grafted and MAH grafted respectively) were mixed in a high speed mixer at 75°C for 15 min, then compounded in a co-rotating twin screw extruder to produce homogeneous composites and cut into granules, and finally they were extruded molding in a single screw extruder through a rectangular die with the dimensions of 4 mm×40 mm. The processing temperature of extrusion was set at the temperature range of 150–180 °C for different zones. The rotary speed of screw was 50 rpm for twin screw extruder and 10 rpm for single screw extruder.

Rheological tests

Rheological properties of composites were measured by an AR2000ex rheometer (TA Instrument, USA) using a 25-mm stainless steel parallel plate. The dynamic oscillatory strain sweep test was undertaken at the strain amplitude of 0.001%–100%. The oscillatory frequency sweep was from 100 to 0.01 rad/s for the composites at 190°C, and the constant strain amplitude of 0.01% was applied, which was well within the linear viscoelastic region (LVR).

Mechanical tests

The flexural test was carried out on a RGT-20A universal testing machine (Shenzhen, China). The test method and procedure were in accordance with the standard test method for flexural properties of unreinforced and reinforced plastics (ASTM D 790-03). The dimensions of the specimens were approximately 80 mm×13 mm×4 mm with a span length of 64 mm. A crosshead speed of 2 mm/min was used when composites were tested. The measurements were performed at ambient conditions, i.e., a temperature of 20 °C and a relative humidity of approximately 50%. At least, 5 specimens of each sample were tested to obtain the flexural modulus and flexural strength.

The impact properties were determined by using a XJ-50G impact tester (Shenzhen, China) according to the standard test method for plastic (GB/T 1043-1993). The dimensions of the specimens were approximately 80 mm×10 mm×4 mm with a span length of 60 mm. The velocity of the tests was set at 2.9 m/s, and the impact energy was 2J. The measurements were performed at ambient conditions, i.e., a temperature of 20°C and a relative humidity of approximately 50%. At least, 5 specimens of each sample were tested to obtain the impact strength.

Results and discussion

Rheological properties

The nonlinear feature gives birth to a few issues about the rheology of the wood plastic composites melts. Thus, it is essential to determine the linear viscoelastic region (LVR) as the first step.

The LVR of the composites with different MAH dosages was determined by strain sweep test. The storage modulus (G') showed especial dependence on the strain (Fig. 1). At the low strain (<0.1%), the G' is independent of strain, which exhibits the linear viscoelastic behavior of composites melt. When the strain is more than 1%, the nonlinear behavior is further evident. The effect of the MAH dosage on the LVR is not evident, but the G' varies with the MAH dosages, especially the G' of 1.5 wt.% MAH is lower than that of other MAH dosages.

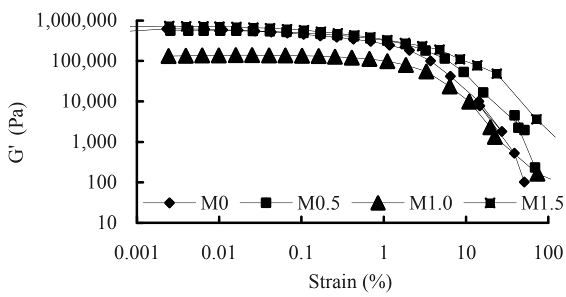

Fig. 1 Strain dependence of the storage modulus (G') of composites with different MAH dosages

Taking account of the LVR observed in the strain sweep test, a strain of 0.01% was selected for the frequency sweep test. The frequency dependence of the storage modulus (G') of composites with different MAH dosages was illustrated (Fig. 2). It can be seen that the G' of the MAH modified composites was lower than that unmodified. When the MAH dosage was 1.5 wt.% (M1.5),

the frequency dependence of the G' was decreasing, especially at low frequency region. It is indicated that the low molecular weight segments, which were generated during grafting procedure, made the chains easy to slide from each other in the melt. However, with the further increment on the MAH dosage, the interactions between anhydride and hydroxyl of the wood fiber are much stronger, which leads to the greater rigid of the composites.

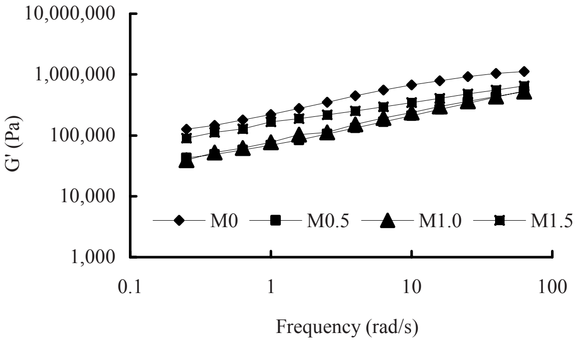

Fig. 2 Frequency dependence of the storage modulus (G') of composites with different MAH dosages

All the composites exhibited shear thinning behavior (Fig. 3), which could be explained that with the increasing of the frequency there was not enough time for the polymer chains to reform the original filler particle distribution (Marcovich et al. 2004). The complex viscosity (η*) of the MAH modified composites was decreased comparing to that of the unmodified composites. A possible explanation why the η* of the MAH modified composites was decreased, was that the matrix molecular weight was decreased during grafting procedure. When the MAH dosage reached a certain value, the chain mobility was restricted by dense polar groups, thus the η* of the composites with M1.5 increased.

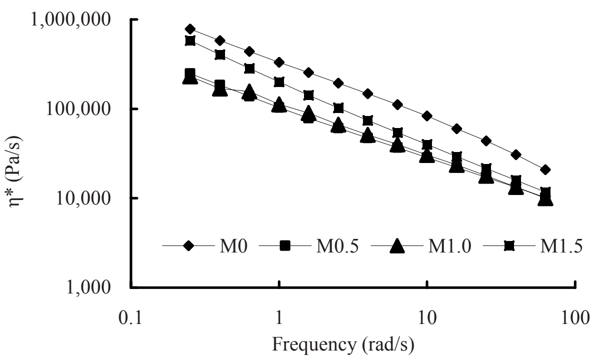

Fig. 3 Frequency dependence of the complex viscosity (η*) of composites with different MAH dosages

Mechanical properties

Mechanical properties at each experimental condition were determined and the values of various properties were plotted against MAH dosage in Figs. 4–6. The influences of different MAH dosages on the flexural strength, flexural modulus and impact strength of the composites were discussed below.

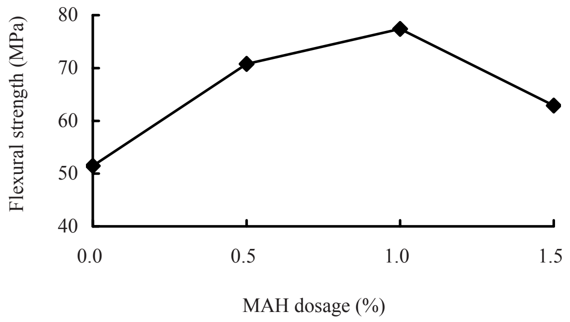

Fig. 4 Influence of MAH dosage on the flexural strength

The flexural strength of the composites varied markedly with MAH dosages (Fig. 4). With modification, the flexural strength reached a maximum at 1.0 wt.% of MAH dosage and then decreased with an increase in the MAH dosage. During the flexural test, one side of the specimen is extended and the other side is compressed under loading. Therefore, the extent of fiber distribution and moderate wetting could affect the flexural properties (Balasuriya et al. 2001). For the MAH modified composites, the viscosity of the matrix was decreased due to the side reaction in the course of the grafting. Thus, the matrix flows well enough to get the uniform distribution of the wood fiber and adequate wetting. As MAH dosage increasing, the chain mobility of the matrix is restrained, so the wood fiber is not adequate wetting, which induces the agglomeration of the wood fiber. Thus, it exhibits the decrease of the flexural strength.

The flexural modulus of the composites decreased and reached a minimum at 0.5 wt.% of MAH dosage, and then increased with an increase in the MAH dosage (Fig.5). This trend is not in accordance with the previous researches, which indicates that the addition of MAPP coupling agent can improve the orientation and dispersion of wood fiber in the matrix and enhance the flexural modulus of the composites (Bengtsson et al. 2007).

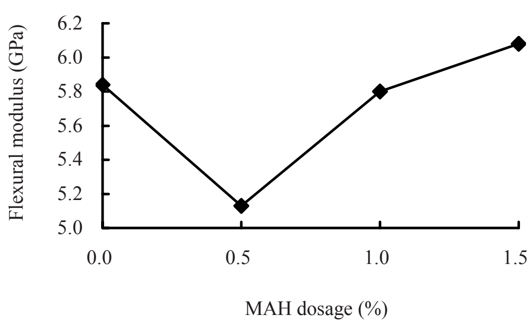

Fig. 5 Influence of MAH dosage on the flexural modulus

This may be attributed to the reduction of the molecular weight which was brought by the degradation reaction during grafting procedure. With the MAH dosage increasing, the grafting reaction becomes dominant and the interactions between the matrix and wood fibers increased. It is known that strong interactions can cause a stiffening effect on the polymer matrix adjacent

to the filler particles interface (Nunez et al. 2003). So the interactions can enhance the interfacial adhesion, which leads to an increase in modulus.

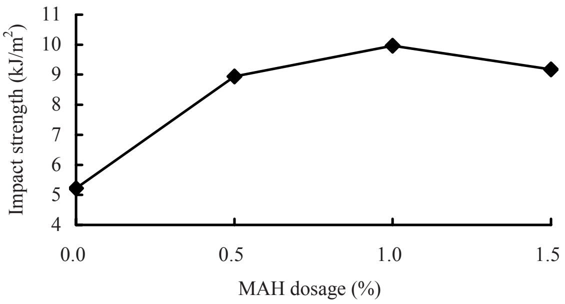

Fig. 6 Influence of MAH dosage on the impact strength

The general trend of the impact strength was the same as that of the flexural strength (Fig. 6), and the impact strength value also reached a maximum MAH dosage at 1.0 wt.%. However, with further increasing the MAH dosage (>1.0 wt.%), the impact strength becomes decreasing. This is consistent with the results reported in most studies (Oksman et al. 1998), which indicated that a weak interface would provide a site for failure. By improving the interfacial adhesion between the matrix and the fibers, the impact strength can thus be improved. In the composites without modification, the wood fibers were randomly distributed in a continuous thermoplastic matrix and encapsulated or enveloped by the thermoplastic matrix mainly with a mechanical connection. For the modified composites, the wood fibers were combined with the thermoplastic matrix through the covalent bonding or hydrogen bonding, thus resulting in a stronger interfacial adhesion (Lu et al. 2005). However, the existence of excessive anhydride groups might enlarge the gap between the wood fiber and thermoplastic matrix and weaken the interface. The loose and weak interfacial connections will act as stress concentrators in the composites, which results in the reduction in the impact strength.

Conclusions

The rheological properties of the resultant composites were studied using dynamic measurement. According to the strain sweep test, the linear viscoelastic region of the composites in the melt was determined. The results showed that the G' was independent of the strain at low strain region (<0.1%). Frequency sweep results indicated that all composites exhibited shear thinning behavior, and the G' and η*of the MAH modified composites were lower than that of the unmodified composites. A further investigation on the different filling systems will help us optimize the process technology, and also reveal the effect of the fillers on the flow performance.

The addition of MAH resulted in the improvement of mechanical properties. The flexural and impact strength of the resultant composites significantly increased and reached a maximum MAH dosage at 1.0 wt.%. After an initial decreased, the flexural modulus of the composites also increased with MAH. The increase in mechanical properties provided evidence of increased interfacial adhesion between wood fiber and modified PP/PE blends. This method can also be considered as a possibility to expand the use of recycled plastics in the manufacture of WPCs.

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